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Chemical composition of radiolytically formed particles using single-particle mass spectrometry

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Abstract

Radiolytic particle formation - the formation of aerosol particles in initially particle-free air through irradiation with ionizing radiation – can be a useful process for the detection of gaseous contaminants in clean air, or an undesirable source of particles in experimental setups. In this work, we observed radiolytic particle formation inside a soft x-ray irradiation chamber in filtered ambient air, but also in active-carbon-filtered synthetic air and nitrogen (>99.999% purity). The largest of the radiolytic particles grew to several 100 nm in diameter. A single-particle aerosol mass spectrometer (LAAPTOF) was used to conduct the first direct measurement of the chemical composition of individual radiolytic particles. Total sample amounts as small as 0.7 ng were analyzed. The mass spectra suggest a chemical composition similar to that of ambient aerosols, with typical secondary species such as nitrate, sulfate, and organics. The strong presence of nitrates is in agreement with previous conclusions on the chemical composition of radiolytic particles, while the presence of organics is shown in this work for the first time. The results show that even very low levels of gaseous organic contaminants in high-purity gases can be detected by virtue of radiolytic particle detection, and that impurities in manufactured gases used must be carefully controlled for in setups where radiolytic particle formation would cause measurement artefacts.

1. Introduction

In radiolytic particle formation, particles are produced from initially particle free air by means of radioactive (e.g. Leong et al. 1983; Ramamurthi et al., 1993, Adachi et al., 1996) or soft X-ray irradiation, for example from soft x-ray aerosol chargers (Yun et al. 2009). Ion-induced nucleation, the presumed initial step of radiolytic particle formation, may also be of atmospheric relevance (e.g. Kirkby et al., 2011 and 2016). Radiolytic particle formation is exploited in a method suggested to detect extremely low levels of gaseous contaminants in clean rooms (airborne molecular contaminants, AMC's), a problem in the semiconductor industry (Kim et al., 2015). In other experimental setups using soft x-ray sources, radiolytic particle formation can be an unwanted side effect. Knowing the size characteristics and the chemical composition of radiolytic particles can help detect such unwanted particle formation, and may indicate which types of trace gases to control for in setups involving soft x-ray sources.

In studies so far, size and concentration of radiolytic particles formed in soft x-ray chargers in filtered ambient air or in controlled mixtures of pure gases were measured. Kallinger et al. (2010a) found that the mean particle diameter increases with the residence time of the gas in the charger, from to 16 nm at a residence time of 20 seconds to 29 nm at a residence time of 2 minutes. Particle number concentrations were found to first increase strongly with residence time (up to 10^6 particles cm^{-3} with a